Circular Dichroism in K-shell Ionization from Fixed-in-Space CO and N_2 Molecules

T. Jahnke[1], Th. Weber[1], A.L. Landers[2], A. Knapp[1], S. Schössler[1], J. Nickles[1], S. Kammer[1], O. Jagutzki[1], L. Schmidt[1], A. Czasch[1], T. Osipov[3], E. Arenholz[4], A.T. Young[4], R. Díez Muiño[5,6], D. Rolles[5], F.J. García de Abajo[5], C.S. Fadley[5,7], M.A. Van Hove[4,5,7], S.K. Semenov[8], N.A. Cherepkov[8], J. Rösch[9], M.H. Prior[9], H. Schmidt-Böcking,[1] C.L. Cocke,[3] and R. Dörner[1]

[1] Institut für Kernphysik, University Frankfurt, August-Euler Str 6, D-60486 Frankfurt Germany
[2] Dept. of Physics, Western Michigan Univ., Kalamazoo, MI 49008, USA
[3] Dept. of Physics, Kansas State Univ, Cardwell Hall, Manhattan, KS 66506, USA
[4] Advanced Light Source, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA
[5] Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA
[6] Donóstia International Physics Center, P. Manuel de Lardizabal 4, 20018 San Sebastián, Spain
[7] Department of Physics, University of California, Davis, CA 95618, USA
[8] State University of Aerospace Instrumentation, 190000 St. Petersburg, Russia
[9] Chemical Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720, USA

INTRODUCTION

This project studies photoemission (photoionization) from "fixed-in-space" molecules, comparing measured angular distributions with theory. In particular, strong circular dichroism in angular distribution (CDAD) is measured for CO and N_2, which is explained by calculations performed both in the multiple-scattering and random-phase-approximation formalisms. The new CDAD measurements, together with earlier results with linearly polarized photons [1,2], constitute the most complete description of K-shell photoemission from a free diatomic molecule. They provide benchmark tests of theoretical methods that are indispensable, for example, in the interpretation of photoemission and photoelectron diffraction data from solids.

METHODS

The present experiment was performed using COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy [3]) at elliptically polarized undulator beamline 4.0.2 of the Advanced Light Source. The resolution for the photoelectron ranges from 1-3 eV depending upon its energy; this is sufficient to discriminate the direct K-shell photoelectron from the Auger and satellite shake up electrons. In the analysis we have used for CO only the $C^+ + O^+$ decay channel with a kinetic energy release KER > 10.2 eV [2], and for N_2 all ions from the $N^+ + N^+$ decay channel.

The calculations for CO are performed in a one-electron model using multiple scattering theory in non-spherical self-consistent potentials (MSNSP) [4] using the experimental values of the electron energy. The molecular ionic potential is split into two touching roughly hemispherical cells in which the full self-consistent potential is present. This allows us to include regions of space neglected by standard multiple scattering theory, and to avoid the usual spherical symmetrization of the potentials around each atomic scattering center. The inclusion of non-spherical effects has been found to be crucial in the calculation of the photoelectron angular distributions for kinetic energies of the electron lower than approximately 30 eV [3].

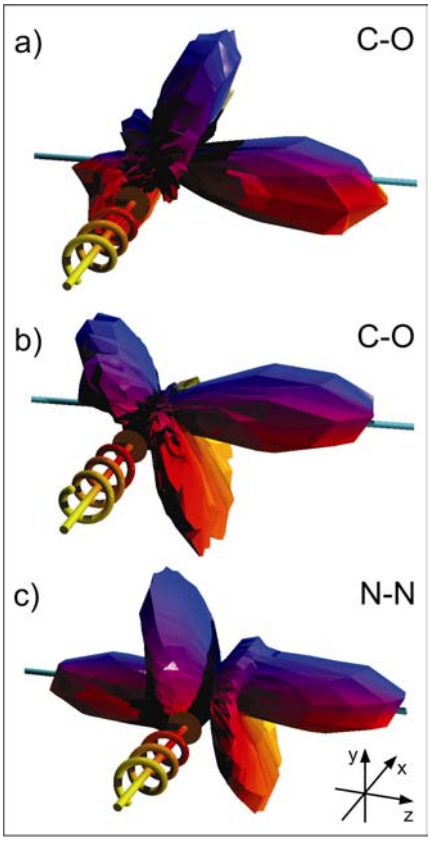

Figure 1. (a) and (b) -- Angular distributions of C(1s) photo-electrons (10 eV kinetic energy, on shape resonance) emitted from a CO molecule by absorption of left and right circularly polarized photons. The sense of rotation of the polarization vector is indicated by the spiral, where the photon propagation vector lies along the + x-axis (i.e., into the page) in all cases. The molecule is aligned along the z-axis, with the carbon atom at negative z in panels (a) and (b). Each vertex of the three-dimensional shape represents one data point. The data have not been smoothed, with the maximum corresponding to about 1000 counts. (c) -- Analogous distribution of N(1s) photoelectrons (9 eV, on resonance) from N_2.

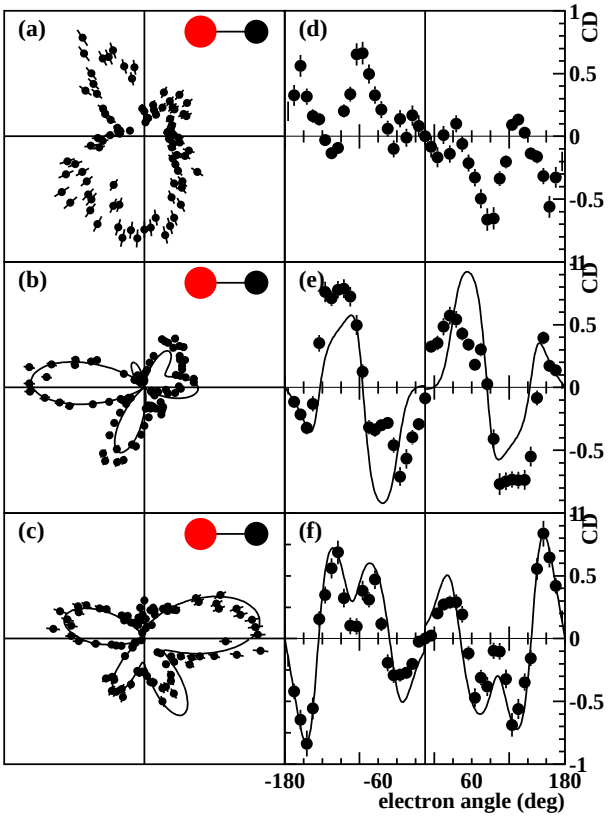

Figure 2. (a-c) Angular distribution of C(1s) photo electrons emitted from a CO molecule by absorption of right circularly polarized photons where the propagation vector of the light is into the page. The molecule lies along the horizontal axis as indicated, and both electrons and molecules lie within 10 degrees of the plane of the page. The electron energies are (a) 1.6, (b) 10.0 and (c) 24.6 eV. Panels (d-f) show the corresponding circular dichroism as defined in the text. Electron angle 0 corresponds to the direction of the carbon. Full lines: Theoretical multiple scattering calculations for the two higher energies, convoluted with the experimental resolution.

For N_2, the calculations take many-electron correlations into account in the Random Phase Approximation (RPA) [5,6]. In this approach, the non-spherical relaxed core Hartree-Fock potential is used as the zero order approximation for the photoelectron wave functions, and the coupling between the 1σ-g and 1σ-u channels is included within the RPA method.

RESULTS

More quantitatively, Fig. 2 shows the photoelectron angular distribution from CO in the y-z-plane (perpendicular to the photon propagation) where CDAD is strongest. The molecule lies along the horizontal axis, as shown in the schematic, and the photon propagation vector is into the page. The electron energies are 1.6, 10.0 and 24.6 eV. The carbon-K σ shape resonance

results in a maximum of the cross section at around 10 eV (306 eV photon energy). The right panels show the CDAD defined as:

$$CDAD = (\sigma_{RCP} - \sigma_{LCP}) / (\sigma_{RCP} + \sigma_{LCP})$$

The theoretical calculations corresponding to the measurements are shown in Fig. 2 by the solid lines, using the multiple-scattering formalism: good overall agreement between experiment and theory is found.

Similar agreement between experiment and theory was obtained (not shown) for N_2, in which case the random-phase-approximation approach was used.

ACKNOWLEDGMENTS

This work was supported in part by BMBF and DFG. R.D. was supported by the Heisenberg Programm der DFG, the Alexander von Humboldt and the Adolf Messer Stiftung. Th.W. thanks Graduiertenförderung des Landes Hessen for financial support. We thank Roentdek GmbH (www.Roentdek.com) for support with the delayline detectors. S.K.S. and N.A.C. acknowledge the financial support of the INTAS-RFBR grant (IR-97-471). R.D. acknowledges many enlightening discussions with J. Berakdar on the relationship of CDAD to phase information.

REFERENCES

1. A. Landers, T. Weber, I. Ali, A. Cassimi, M. Hattass, O. Jagutzki, A. Nauert, T. Osipov, A. Staudte, M.H. Prior, H. Schmidt-Böcking, C.L. Cocke, and R. Dörner, Phys. Rev. Lett. **87**, 013002.
2. T. Weber, O. Jagutzki, M. Hattass, A. Staudte, A. Nauert, L. Schmidt, M. Prior, A. Landers, A. Bräuning-Demian, H. Bräuning, et al, J. Phys. **B34**, 3669 (2001).
3. R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshammer and H. Schmidt-Böcking, Physics Reports **330**, 96 (2000).
4. R. Díez Muiño, D. Rolles, F.J. García de Abajo, F. Starrost, W. Schattke, C.S. Fadley and M.A. Van Hove, J. of Electr. Spectr. and Rel. Phenom. **99**, 114 (2001).
5. S.K. Semenov, N.A. Cherepkov, G.H. Fecher and G. Schönhense, Phys. Rev. **A61**, 032704 (2000).
6. N.A. Cherepkov, S.K. Semenov, Y. Hikosaka, K. Ito, S. Motoki and A. Yagishita, Phys. Rev. Lett. **84**, 250 (2000).

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, Geo-, Bio-, Chemical and Materials Sciences Divisions, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Principal investigator: Michel A. Van Hove, Advanced Light Source and Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory. Email: MAVanHove@lbl.gov. Telephone: 510-486-6160.